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Anomalous Hall Effect in Double Exchange Magnets

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We investigate the possible origin of anomalous Hall effect in the CMR (colossal magnetoresistance) materials - the doped rare earth manganites - observed recently by Matl *et al* [1]. It is demonstrated that the spin-orbit interaction in the double exchange model couples magnetization to the Berry phase associated with three dimensional spin textures and induces a non-zero average topological flux which in turn generates an anomalous contribution to transverse resistivity. The same effect, but involving the orbital Berry phase, occurs in the model with orbital degeneracy and Coulomb repulsion.

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It has been known for a long time [2] that the Hall effect in ferromagnets exhibits a low field "anomaly" which can be parametrized as magnetization, M, dependent contribution to transverse resistivity: $\rho_{xy} = R_H B + R_A M$. The anomalous Hall constant R_A is usually explained in terms of skew scattering due to spin-orbit coupling as reviewed in Ref [2]. Recently Matl *et al* [1] measured the Hall resistivity of $La_{1-x}Ca_xMnO_3$ with $x = 0.3$ and found negative R_A (in contrast with positive, hole like, R_H) which becomes significant above $100K$ and peaks about $20K$ above the Curie temperature $T_c \approx 265K$. This temperature dependence (which follows closely that of the longitudinal resistivity) contrasts with the behavior of, for example, Ni, where R_A peaks well below T_c and contradicts to the existing theories [4,5,15]. There exist also related Hall measurements on similar material at relatively high temperatures [6] and on $La_{1-x}Ca_xCoO_3$ $(0.1 < x < 0.5)$ [7]. These new data lead us to revisit the theory of the anomalous Hall effect.

The Manganites are described by a model which includes double exchange, Jahn-Teller, and electron-electron (Hubbard) interactions:

$$H_0 = -t \sum_{r,\mu} c^{\dagger}_{r,\sigma,a} c_{r+\mu,\sigma,a} - J_{\rm H} \sum_r \mathbf{S}_r \cdot c^{\dagger}_{r,\sigma,a} \hat{\tau}_{\sigma\sigma'} c_{r,\sigma',a} + H_{\rm JT} + H_{e-e} \; . \tag{1}$$

where $c^{\dagger}_{r,\sigma,a}$ is the creation operator for the conduction electron with spin σ and the orbital index $a = 1, 2$, $t \sim 0.1 - 0.5eV$ is the hopping matrix element and $\mathbf{S}_r$ is the core spin. This model is suggested by the electronic structure of the doped rare earth manganites [8–10] ($Re_{1-x}A_xMnO_3$ where Re is a rare-earth element such as La or Nd and A is a divalent metal ion such as Sr or Ca) where $1 - x$ electrons occupy the orbitally degenerate e_g states ($d_{x^2-y^2}$ and $d_{3z^2-r^2}$) and couple strongly to the core $S = 3/2$ moment via Hund's rule, $J_H \sim 1 - 2eV$. The on-site Coulomb repulsion $H_{e-e} = U \sum_r n_{r,\uparrow,a} n_{r,\downarrow,a} + V \sum_{r,\sigma,\nu} n_{r,\nu,1} n_{r,\sigma,2}$ (where $n_{r,\sigma,a} \equiv c^{\dagger}_{r,\sigma,a} c_{r,\sigma,a}$) is believed to be of the same order as J_H [11,12]. In addition, it has been argued [13,14] that in the rare earth manganites an important role is played by the Jahn-Teller (JT) coupling of the e_g electron with the phonon mode $\mathbf{Q}$ describing a local tetragonal distortion which lifts orbital degeneracy via $H_{\rm JT} = g \sum_r c^{\dagger}_{r,\sigma,a} \mathbf{Q}^{ab}_r c_{r,\sigma,b}$. This coupling can explain the transition from metallic to activated transport at T_c for a certain range of doping x. Below we shall discuss both the case of large local distortions where the orbital degree of freedom is essentially quenched and the case where distortions are weak and the orbital degeneracy is retained (at least on the scale of k_BT) which may be appropriate for the ferromagnetic metal phase of the compounds in question.

Since the anomalous Hall effect manifests itself as the dependence of ρ_{xy} directly on the magnetization, it must originate from the spin-orbit interaction [2]. It includes the (relativistic) interaction of the spin of the itenerant electron with the local electric field and the magnetic interaction of the itinerant electron with the core spins. The latter term, $\mathbf{S} \cdot \mathbf{p} \times \mathbf{r}/r^3$, because spin-orbit coupling is quenched by the cubic crystal field has no matrix element bewteen e_g electrons on the same site, but it does have matrix elements bewteen different sites. In the continuum limit, the sum of both contributions can be written as [15,16]

$$H_{\rm so} = i \int d^3r \; u_{ab} \; \epsilon_{ijk} \; S_i(r) \; \partial_j c^{\dagger}_{\sigma,a}(r) \; \partial_k c_{\sigma,b}(r) \; . \tag{2}$$

As with the hopping matrix, we shall for simplicity ignore the orbital dependence of the coupling constant and take $u_{ab} = \delta_{ab} u$. With the spatially uniform spin-orbit interaction [17] the coupling to the uniform component of

$\mathbf{S}(r)$, *i.e.*, magnetization, has the form of a total derivative and can be integrated out to the boundary. Yet, we shall now show that in the presence of interactions this *topological* term is important and generates the anomalous Hall effect. Previous authors have studied spatially non-uniform spin configurations due to magnetic impurities or spin-wave excitations about the ferromagnetic ground state. Here we argue that Berry phase effects caused by non-coplanar spin configurations give the dominant contribution to ρ_{xy}, at least in the limit of strong carrier-spin couplings appropriate to the maganites.

As a mathematical convenience to make the Berry phase physics explicit, we use the standard Hubbard-Stratonovich approach in which we express the interacting electron in terms of a non-degenerate spinless fermion ψ_r and the slave boson fields $z_{r,\sigma}$ and $w_{r,a}$ which parametrize the spin and the orbital degrees of freedom respectively: $c_{\sigma,a} = \psi^\dagger z_\sigma w_a$. The bosons obey the single occupancy constraint: $\sum_\sigma \bar{z}_{r,\sigma} z_{r,\sigma} = \sum_a \bar{w}_{r,a} w_{r,a} = 1$ and the average fermion density is $\langle \psi_r^\dagger \psi_r \rangle = x$. Neglecting terms of relative order $t/J_{\rm H}$ and t/U, we find

$$H_{\rm eff} = t \sum_{r,\mu} \mathbf{z}_{r+\mu}^\dagger \mathbf{z}_r \mathbf{w}_{r+\mu}^\dagger \mathbf{w}_r \psi_r^\dagger \psi_{r+\mu} - J_{\rm H} \sum_r \mathbf{S}_r \cdot \mathbf{z}_r^\dagger \hat{\tau} \mathbf{z}_r \psi_r \psi_r^\dagger + g \sum_r \mathbf{w}_r^\dagger \mathbf{Q}_r \mathbf{w}_r \, \psi_r \psi_r^\dagger \ , \tag{3}$$

where $\mathbf{z}^\dagger = (\bar{z}_\uparrow, \bar{z}_\downarrow)$ and $\mathbf{w}^\dagger = (\bar{w}_1, \bar{w}_2)$. The factorization of $c_{\sigma,a}$ not only enforces single occupancy but projects onto the low energy spin and orbital state. Assuming $J_{\rm H} \gg t$ the carrier spin and hence $\mathbf{s} \equiv \mathbf{z}_r^\dagger \hat{\tau} \mathbf{z}_r$ is aligned with $\mathbf{S}_r$. The generally complex spin overlap factor $\mathbf{z}_{r+\mu}^\dagger \mathbf{z}_r$ is then determined by the Euler angles of the corresponding local moments and is identical to the one found in a conventional derivation of double exchange [10]. Orbital degree of freedom results in an analogous *orbital* overlap factor $\mathbf{w}_{r+\mu}^\dagger \mathbf{w}_r$. Note however that since the tetragonal/orthorhombic phonon field $\mathbf{Q} = Q_1 \tau^x + i Q_2 \tau^y$ is real, large $g|\mathbf{Q}|$ forces the orbital "isospin" $\mathbf{n} \equiv \mathbf{w}^\dagger \hat{\tau} \mathbf{w}$ into planar configurations. We emphasize that although this formalism is the most natural one in the strong coupling limit, we expect the physics to apply at any coupling, with possibly different amplitude as discussed below.

The Hamiltonian (3) is invariant under *two* local $U(1)$ transformations [12,18]. One of them involves the spin degree of freedom and is given by $\psi_r \to \psi_r e^{i\xi_r}$ and $\mathbf{z}_r \to \mathbf{z}_r e^{i\xi_r}$. This spin $U(1)$ symmetry is broken in the ferromagnetic (FM) phase, but is restored in the paramagnetic (PM) phase. The other one involves the orbital degree of freedom and represented as $\psi_r \to \psi_r e^{i\zeta_r}$ and $\mathbf{w}_r \to \mathbf{w}_r e^{i\zeta_r}$. As long as the orbital ordering does not occur [13,12] the orbital $U(1)$ symmetry is preserved. These $U(1)$ symmetries give rise to two gauge fields which are defined as

$$\begin{aligned} a_\mu^s &= \frac{i}{2}[(\partial_\mu \mathbf{z}^\dagger)\mathbf{z} - \mathbf{z}^\dagger \partial_\mu \mathbf{z}] \\ a_\mu^n &= \frac{i}{2}[(\partial_\mu \mathbf{w}^\dagger)\mathbf{w} - \mathbf{w}^\dagger \partial_\mu \mathbf{w}] \ . \end{aligned} \tag{4}$$

Non-trivial configurations of the gauge field are generated by non-coplanar arrangements of spin or orbital moments; they affect carrier motion just as does an external magnetic field, and may therefore give rise to an anomalous Hall effect. Their coupling to the charge, spin, and orbital degrees of freedom is particularly transparent in the continuum limit [19,20] of (3):

$$H_{\rm cont} = \int d^3 r \ [t_{\rm eff} \psi^\dagger (i\partial_\mu + eA_\mu + a_\mu^s + a_\mu^n)^2 \psi \ + \rho_{\rm eff}^s \mathbf{z}^\dagger (i\partial_\mu + a_\mu^s)^2 \mathbf{z} + \rho_{\rm eff}^n \mathbf{w}^\dagger (i\partial_\mu + a_\mu^n)^2 \mathbf{w} + g \mathbf{w}^\dagger \mathbf{Q}(r) \mathbf{w}] \tag{5}$$

where $t_{\rm eff}$ is the effective hopping parameter of charge carriers and the spin and orbital stiffness $\rho_{\rm eff}^{s,n}$ are defined in the sense of a self-consistent mean field theory. $\mathbf{Q}(r)$ is the quenched random phonon field. The scale of $\rho_{\rm eff}^{s,n}$ is set by the electron kinetic energy while the effective hopping of the holes at finite temperatures is reduced from its bare value t by the fluctuation of the local moments and the orbital degree of freedom. The $\langle \psi \psi^\dagger \rangle = 1 - x$ factor appearing in the MFT, can be absorbed into resetting the boson density $\mathbf{z}^\dagger \mathbf{z} = \mathbf{w}^\dagger \mathbf{w} = 1 - x$. We have introduced the electromagnetic gauge field $\mathbf{A}$ which only couples to the charge of the fermion ($e > 0$ since $\psi^\dagger$ creates holes). The key point in (5) is that the fermions feel an effective magnetic field $\mathbf{B}_{\rm eff} = \mathbf{B} + e^{-1}\mathbf{b}^s + e^{-1}\mathbf{b}^n = \nabla \times (\mathbf{A} + e^{-1}\mathbf{a}^s + e^{-1}\mathbf{a}^n)$ which includes the contribution of the bosonic Berry phases described by $\mathbf{a}^{s,n}$.

The spin-orbit Hamiltonian (2) includes the coupling to the Berry phase which is made explicit by substituting $c_{\sigma,a} = \psi^\dagger z_\sigma w_a$.

$$H'_{\rm so} \approx \int dr^3 \, u \langle \psi \psi^\dagger \rangle \, \mathbf{M} \cdot \nabla \times (\mathbf{a}^n + \mathbf{a}^s) \tag{6}$$

where we have assumed that $\mathbf{z}$ and $\mathbf{w}$ are slowly varying functions and suppressed the terms with derivatives acting on ψ. We observe here that the spin-orbit interaction couples the average magnetization to the bosonic Berry phases

(topological flux) $\mathbf{b}^{s,n}$. The latter arises through non-planar configurations of the $\mathbf{s}(r)$ and/or $\mathbf{n}(r)$ via $\mathbf{b}^s_i(r) = \epsilon_{ijk}\mathbf{s}\cdot\partial_j\mathbf{s}\times\partial_k\mathbf{s}$ and analogously for $\mathbf{b}^n$. Note however that strong Jahn-Teller effect (*i.e.* $g\,|\mathbf{Q}| \gg k_BT$) would confine $\mathbf{n}$ to a plane and suppress $\mathbf{b}^n$. The fact that the spin-orbit interaction couples magnetization to the topological flux associated with spin (or orbital) textures is quite general and not restricted to the strong coupling limit and any slave boson/fermion parametrization. However, the effects become weak for small $J_{\rm H}/t$.

Let us start by considering the case of quenched orbital phases, $\mathbf{b}^n = 0$ and analyse the effect of spin textures in the paramagnetic and ferromagnetic phases. In the FM phase $\mathbf{z}$ bosons are Bose-condensed corresponding to FM order in $\mathbf{s}$. As a result, the spin gauge field $\mathbf{a}^s$ becomes massive with the mass proportional to the density of the condensate and thus there is no uniform contribution to $\mathbf{b}_s$. On the other hand in the PM phase we expect to find a quadratic contribution, $(\mathbf{b}^s)^2$, to the total energy and a finite topological susceptibility defined as $\chi_T \equiv \partial\langle b^s\rangle/\partial M$. The calculation proceeds by integrating out the ψ and $\mathbf{z}$ fields in Eq.(5) and results in an effective Hamiltonian:

$$H_b = \int d^3r\,[\chi_F\ (e\mathbf{B}\,+\,\mathbf{b}^s)^2 +\ \chi_B\,(\mathbf{b}^s)^2 + u(1-x)\ \mathbf{M}\cdot\,\mathbf{b}^s] \tag{7}$$

with the diamagnetic susceptibilities $\chi_F = \pi^{-2}t^2_{\rm eff}D(\epsilon_F)$ and $\chi_B = n(0)\sqrt{2T\rho^s_{\rm eff}}$ (where $n(\epsilon) = 1/(e^{(\epsilon-\mu_B)/T} - 1)$ is the occupation number of the $\mathbf{z}$ bosons) are determined by the current/current correlators of the fermions and bosons respectively. Minimizing with respect to $\mathbf{b}^s$ yields

$$\langle\mathbf{b}^s\rangle\ = -\frac{u(1-x)}{2(\chi_F+\chi_B)}\mathbf{M} - \frac{e\chi_F}{\chi_F+\chi_B}\mathbf{B} \tag{8}$$

from which we read off the topological susceptibility $\chi_T = -\frac{u(1-x)}{2(\chi_F+\chi_B)}$. Magnetization here refers to the average local moment in units of $g_e\mu_BS$ where g_e is the Lande g-factor and μ_B is the Bohr magneton. The topological flux per plaquette of the lattice is of order $u/\chi_{F,B}$ which we can estimate conservatively by taking $u \sim 1-10K$ and $\chi \sim t$ yielding $\langle\mathbf{b}^s\rangle \sim 10^{-3} - 10^{-2}l^{-2}$ where l is the lattice constant. This flux is comparable to $eB/\hbar c$ for $1T$ field. One also observes that the flux is maximized for a nearly full band as may be expected for the effect arising from interactions.

According to the Ioffe-Larkin phenomenology [21] the constraint between the fermionic and the bosonic currents which follows from (3) implies that the Hall resistivity ρ_{xy} is given by the sum of fermionic ρ^F_{xy} and the bosonic ρ^B_{xy} contributions. Using the semiclassical Hall constants corresponding to fermion density $n_F = x$ and boson density $n_B = 1-x$, we have $\rho^F_{xy} = (eB + b^s)/e^2n_F$ and $\rho^B_{xy} = b^s/e^2n_B$ so that

$$\rho_{xy} = -\frac{u(1-x)(n_F^{-1}+n_B^{-1})}{2e^2(\chi_F+\chi_B)}\,M + \frac{\chi_Bn_F^{-1} - \chi_Fn_B^{-1}}{\chi_F+\chi_B}\,\frac{B}{e}\ . \tag{9}$$

from which we identify:

$$\begin{aligned} R_H &= \frac{1}{e}\frac{\chi_Bn_F^{-1} - \chi_Fn_B^{-1}}{\chi_F+\chi_B} \\ R_A &= -\frac{u(1-x)(n_F^{-1}+n_B^{-1})}{2e^2(\chi_F+\chi_B)} \end{aligned} \tag{10}$$

According to the naive classical estimate, $u > 0$ and R_A has the sign [22] opposite to R_H. This could explain the sign of the anomalous Hall effect observed by Matl *et al* [1]. Note also the compensation of R_H by the bosonic contribution: the large apparent carrier concentration observed in the experiment suggests that some compensation effect does indeed exist. However, the calculation of the Hall constant in an interacting system clearly requires greater effort than the semiclassical estimates.

The above estimate for χ_F given above is appropriate for the metallic paramagnetic phase (such as observed in the doped rare earth manganites at the values of x somewhat higher than optimal value for the CMR effect); at lower dopings the PM phase has very high resistivity, thus we expect in this situation that $\chi_F \ll \chi_B$ ($D(\epsilon_F) \to 0$) and the relevant carrier density n_F ($n_F^{-1} \gg n_B^{-1}$) to be thermally activated. For the CMR compounds, the insulating PM phase is likely to be caused by the large local Jahn-Teller distortions [13,14] which split the conduction band into filled orbitally non-degenerate subband with spectral weight $1-x$ and an empty orbitally degenerate subband with spectral weight x. Conduction is due to the thermally excited carriers in both subbands and the proper determination of the sign and magnitude of R_H is non-trivial. (Alternatively one may think of transport in terms of small polarons [6].) However the appearance, due to spin-orbit interaction, of the average topological field $\langle\mathbf{b}^s\rangle$ as well as its coupling to the carriers are robust. We expect $R_A \approx e^{-1}\chi_TR_H$ with $\chi_T = -u(1-x)(2\chi_B)^{-1}$, which follows from (10) under

$\chi_F \ll \chi_B$ and $n_F^{-1} \gg n_B^{-1}$ assumptions but can be understood simply by saying that $\langle \mathbf{b}^s \rangle$ acts just like magnetic field.

Let us now discuss the ferromagnetic phase. As already noted, in the FM phase the $\mathbf{a}^s$ gauge field acquires a mass due to the divergence of χ_B at $k=0$. This phenomenon is reminiscent of the Meissner effect in a superconductor, but there is a crucual difference due to the fact that the topological flux on the lattice is not conserved. A continuum FM spin texture with $\mathbf{b}_i^s(r) = \epsilon_{ijk}\mathbf{s}\cdot\partial_j\mathbf{s}\times\partial_k\mathbf{s} \neq 0$ is the Belavin-Polyakov [23] skyrmion which in a 3D ferromagnet would be a line defect (akin to a flux tube) with the line tension $2\pi\rho^s_{\text{eff}}|q|$ where q is the topological charge equal to the integral of $\mathbf{b}^s$ over the crossection of the "tube" divided by 8π. Yet when this continuum solution is transfered to the lattice, the quantization of $|q|$ is lost since shrinking the lateral size of the skyrmion allows the texture to "fall through" the plaquette. For the same reason (and in contrast to a flux line in a superconductor) the skyrmion line can have free ends and at $T \neq 0$ there will be a finite density of such objects. Alternatively one can argue that the spins around any plaquette may be deformed into a texture carrying topological flux $q = \frac{l^2 b^s}{8\pi}$ at an energy cost $\alpha\rho^s_{\text{eff}}|q|$ (α being a constant of $O(1)$) which should be contrasted with the quadratic energy cost in the PM. One can calculate the topological susceptibility from the partition function $Z(M) = \int d\mathbf{b}^s \exp[-(\alpha\rho^s_{\text{eff}}/8\pi)|\mathbf{b}^s|/T - u\mathbf{b}^s\cdot\mathbf{M}]$. One finds $\chi_T \approx -8\pi uT/\alpha\rho^s_{\text{eff}}$ for $T/\rho^s_{\text{eff}} \ll 1$. Since the topological $\mathbf{b}$ couples to the carriers exactly like the magnetic field we expect: $R_A = e^{-1}\chi_T R_H$. Note that approaching T_c the topological susceptibility χ_T should be enhanced both by the softening of ρ^s_{eff} (due to the reduction of electron kinetic energy [13] which underlies the FM exchange constant) and by the contribution of the large scale skyrmion textures which take advantage of the vanishing spin stiffness at long wavelength. The enhancement of χ_T and hence R_A could explain the observed upturn of R_A below T_c.

While the temperature dependence of R_A indicated by the above arguments is qualitatively consistent with that observed by Matl *et al* [1] the region close to T_c requires a more careful treatment. At present we neither understand why (or whether) $R_A(T) \sim \rho(T)$ nor why both quantities peak $20-30K$ *above* T_c. An *a priory* calculation of R_H is beyond our present scope. However we did demonstrate how in the presence of interactions the spin-orbit coupling can generate direct dependence of ρ_{xy} on magnetization via the appearance of topological flux. This should be contrasted to the traditional view [4,5] of skew-scattering contributing to ρ_{xy} in the $3d$ order via $\langle(M-\langle M\rangle)^3\rangle$ which peaks well below T_c.

Curiously, the analysis presented for the PM phase applies equally well to the description of the orbital Berry phase effects in the orbital "liquid" state [12] ($g|\mathbf{Q}| \ll k_B T$) where the orbital degeneracy is restored in FM phase. While in double exchange problem the Hubbard U played no role as the single occupancy was ensured by the Hund's rule splitting, if the orbital degeneracy is restored, the effect of U becomes important and necessary for the appearence of the topological effects. In this case, $\mathbf{b}^s$ in Eq.7 and Eq.8 is replaced by $\mathbf{b}^n$ and the rest of the discussion goes in the same way. Therefore, if U is large and $g|\mathbf{Q}|$ small, the effect of the orbital Berry phase would provide additional contribution to the anomalous Hall coefficient in FM phase.

In conclusion we emphasize the generality of the physical mechanism by which the topological textures in magnetic systems get polarized by the magnetization via spin-orbit coupling and contribute to ρ_{xy}. Comparing an itenerant magnet to the double exchange magnet considered above, the former corresponds to a weak coupling regime, where the local moments and hence any possible textures with $\langle \mathbf{S}_i\cdot\mathbf{S}_j\times\mathbf{S}_k\rangle \neq 0$ disappear at T_c. By contrast, manganites with their core spins and large J_H are in the strong coupling limit. While our work points to the possible origin of the unusual anomalous Hall effect in doped manganites much further work is required to make a quantitative comparison. It would also be interesting to measure the anomalous Hall effect for $\mathrm{La_{0.7}Sr_{0.3}MnO_4}$ or the like compound, which remains metallic in the PM phase, which could be compared with our result on PM metallic phase.

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